LASER MODULATION AT THE ATOMIC LEVEL

Monthly Report No. 3

Date of this report: 10 October 1964
Period covered: 1 September 1964 to 30 September 1964

Submitted to
National Aeronautics and Space Administration
Contract No. NASw 1008

GENERAL DYNAMICS | ELECTRONICS

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LASER MODULATION AT THE ATOMIC LEVEL

Purpose

Research on methods of influencing internally the radiating centers of active laser materials in order to achieve laser modulation is the principal objective of the work carried out under this contract.

Summary

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The absorption of a ruby rod is described in the presence of absorption by the metastable state in the wavelength region of interest and with some inhomogeneity in pumping along the length of the rod. The results of transmission measurements at room temperature and at 115°K are presented and interpreted by means of the theory. Initial testing of a YAG: Nd^{3+} laser rod is described.

Man-Hours Worked

The total number of man-hours worked during the reporting period is 892.5 hours.

I. STUDY OF RUBY

A. Theoretical Discussion of the Transmission of Light through Ruby in the Presence of Excited State Absorption

As explained in Monthly Report No. 2, it has been found necessary to use somewhat more sophisticated assumptions concerning our experimental system than those described in Monthly Report No. 1. We now take into account both absorption of radiation by ions in the metastable ^2E levels and inhomogeneity of pump radiation along the length of the rod. The latter effect is due primarily to the copper loop around the center of the rod used in generating the magnetic field.

For the purpose of the present discussion we define I_o as the intensity of a light beam inside the ruby surface at x = 0, and I(x) as the intensity at the plane x inside the ruby. We therefore need consider only bulk absorption, and not surface scattering or reflection. For a homogeneous rod extending from x = 0 to x = L the intensity of a transmitted light beam I_L is given by:

$$\int_{I_o}^{I} \frac{dI}{I} = \ln \frac{I(L)}{I_o} = - \int_0^L \sum_{i,j} \sigma_{i,j}(\lambda)(n_i - n_j)\, dx , \qquad (1)$$

where $\sigma_{i,j}$ is the absorption cross section for the transition from state i to state j, and n_i and n_j are the populations of states i and j, respectively. The summation is carried out in such a way that j > i in all terms, the states being numbered in order of increasing energy. For the case of Cr^{3+} in ruby, the only excited levels which achieve appreciable population densities under our pumping conditions are those belonging to the ^2E state that gives rise to the R_1 and R_2 fluorescence lines. We can therefore modify Eq. (1) by letting i represent only components of the $^4A_2$ ground state, j represent those of the ^2E state, and k represent all higher excited states, obtaining:

$$\ln \frac{I}{I_o} = - \int_o^L [\sum_{i,j} \sigma_{i,j} (n_i - n_j) + \sum_{i,k} \sigma_{i,k} n_i + \sum_{j,k} \sigma_{j,k} n_j]\, dx . \qquad (2)$$

For the most part the higher states are rather broad bands which do not permit resolution of the contributions of the separate lower state components to the cross section. This permits us to write:

$$\frac{1}{n} \sum_{i,k} \sigma_{i,k} n_i = \sigma_o$$

and

$$\frac{1}{n_m} \sum_{j,k} \sigma_{j,k} n_j = \sigma^* ,$$

where n is the total ground state population and n_m is the total ^2E state population, σ_o and σ^* being the ground and excited state absorption cross sections respectively, using the notation of Gires and Meyer.[1] In regions of the spectrum not coincident with the R lines, the transmitted intensity is therefore given by the rather simple relation

$$\ln \frac{I}{I_o} = - \sigma_o \int_0^L n\, dx - \sigma^* \int_0^L n_m dx$$

or, since $n + n_m \simeq N_o$ under these conditions,

$$- \ln \frac{I}{I_o} = \sigma_o N_o L + (\sigma^* - \sigma_o) \int_0^L n_m dx . \quad (3)$$

If σ_o and σ^* are both known for a given wavelength, the apparent population density can be found simply by measuring the intensity transmitted by the unpumped crystal I_u and that of the pumped crystal I_p:

$$\frac{1}{L} \int_0^L n_m dx = \frac{\ln(I_p/I_u)}{L(\sigma_o - \sigma^*)} = \bar{n}_m \quad (4)$$

In order to confirm the results of Gires and Meyer for ruby at room temperature, and in order to obtain the excited state spectrum at low temperature where the magnetic field measurements are carried out, $\sigma^*(\lambda)$ must be determined. From Eq. (4) we see that

$$\sigma^* = \sigma_o - \frac{1}{\bar{n}_m L} \ln \frac{I_p}{I_u} .$$

If transmission measurements are made over a considerable spectral range using constant pump energy,

$$\int_0^L n_m dx = \bar{n}_m L$$

is a constant which can be arbitrarily selected. Arbitrary selection of $\bar{n}_m$ leads to an arbitrary σ^* spectrum which can then be subjected to two types of constraint: (1) $\sigma^* \geq 0$ for all λ and (2) at those wavelengths where $I_p/I_u = 1$, $\sigma^* = \sigma_o$. The latter condition is especially useful with broad spectral bands encountered in ruby in the visible and ultraviolet spectrum, since the points $\sigma^* = \sigma_o$ give absolute calibration points through the σ_o measurements.

1. F. Gires and G. Meyer, "Attenuation et amplification optiques du rubis excite," Quantum Electronics, 1963 Paris Conference, pp. 841 ff, Columbia University Press, New York, 1964.

B. Transmission Experiments

Experimental measurements of σ_o have been reported in Monthly Report No. 2. Spectral measurements of I_p/I_u over the range 4000 Å to 7000 Å have been carried out after continued refinement of the previously described experimental technique. Since the entire "white light" beam from the compact arc lamp pulse is passed through the ruby, it was considered possible that some pumping is accomplished by the probe pulse, which would distort the absorption measurements. This possibility was checked by placing a narrow-band 5400 Å interference filter in front of the laser cavity to reduce unnecessary pump light. The ratio I_p/I_u did not vary by more than the experimental error whether the filter was in or out, so that the assumption of negligible pumping by the probe light is known to be valid.

The excited state absorption cross section was evaluated as described in preceding Section A. Excellent matching could be obtained for all three points at which $\ln I_p/I_u = 0$, even though $\bar{n}_m$ is the only independent variable used, which is a partial confirmation of the validity of the present assumptions. The curves of $\sigma_o(\lambda)$ and $\sigma^*(\lambda)$ obtained from these experiments for $E \perp c$ are shown in Fig. 1 for a 0° ruby (ruby #5) at room temperature. The agreement with Gires and Meyer[1] is quite good if we assume that Fig. 3 of their paper contains a drafting mistake and the curve for $\pi_\omega(E \perp c)$ should cross the curve for $\pi_\epsilon(E \parallel c)$ at 20,000 cm^{-1} (5000 Å). Figure 2 shows the spectral dependence of I_p/I_u, from which σ^* was derived. Figure 3 presents the spectral dependence of I_p/I_u at 115 °K. The data from the low temperature run have not yet been reduced to obtain σ_o and σ^*.

The experimental measurement of $\bar{n}_m$ at laser threshold in the presence of a magnetic field has been tested, and now appears simpler than had been anticipated. Once laser threshold has been reached, increased pumping of the volume which emits laser radiation merely increases radiative output, but does not lead to increased population inversion. If the probe beam is coincident with the volume engaged in laser oscillation, the value of $\ln(I_p/I_u$ or $\bar{n})$, is seen to remain constant for pump energies above threshold. It therefore becomes unnecessary to determine threshold, but merely to insure laser oscillation in the probed volume in order to measure population inversion required to achieve threshold in the presence of a magnetic field. If the probe beam is displaced from the volume of lowest threshold, the saturation of I_p/I_u is noted only when the region of oscillation expands to include the volume sampled by the probe beam. A curve of $\ln I_p/I_u$ versus pump energy taken under such conditions is shown in Fig. 4, the saturation occurring about 40 joules above threshold for laser oscillation because the center of the probe beam was about 2 mm below the laser rod center. All the measurements reported here have been made with an uncoated 0° ruby rod. All laser experiments were performed at about 115° K, where threshold for an uncoated rod is readily achieved.

The determination of σ_0 and σ^* at 115° K and the measurement of $\bar{n}_m$ during laser oscillation in the presence of an inhomogeneous magnetic field for 0° ruby are under way, and similar measurements for the 90° ruby require only that a polarizer be introduced into the transmitted beam to isolate the $E \perp c$ component.

While the experimental results now appear to be very satisfactory, the theoretical calculations are still awaiting solution of the state mixing problem, and also will require some assumptions as to the form of $n_m(x)$. Experimental determination of $n_m(x)$ is very difficult, and it may be necessary to be content with the approximation that n(x) is constant, as has been done in previous calculations.

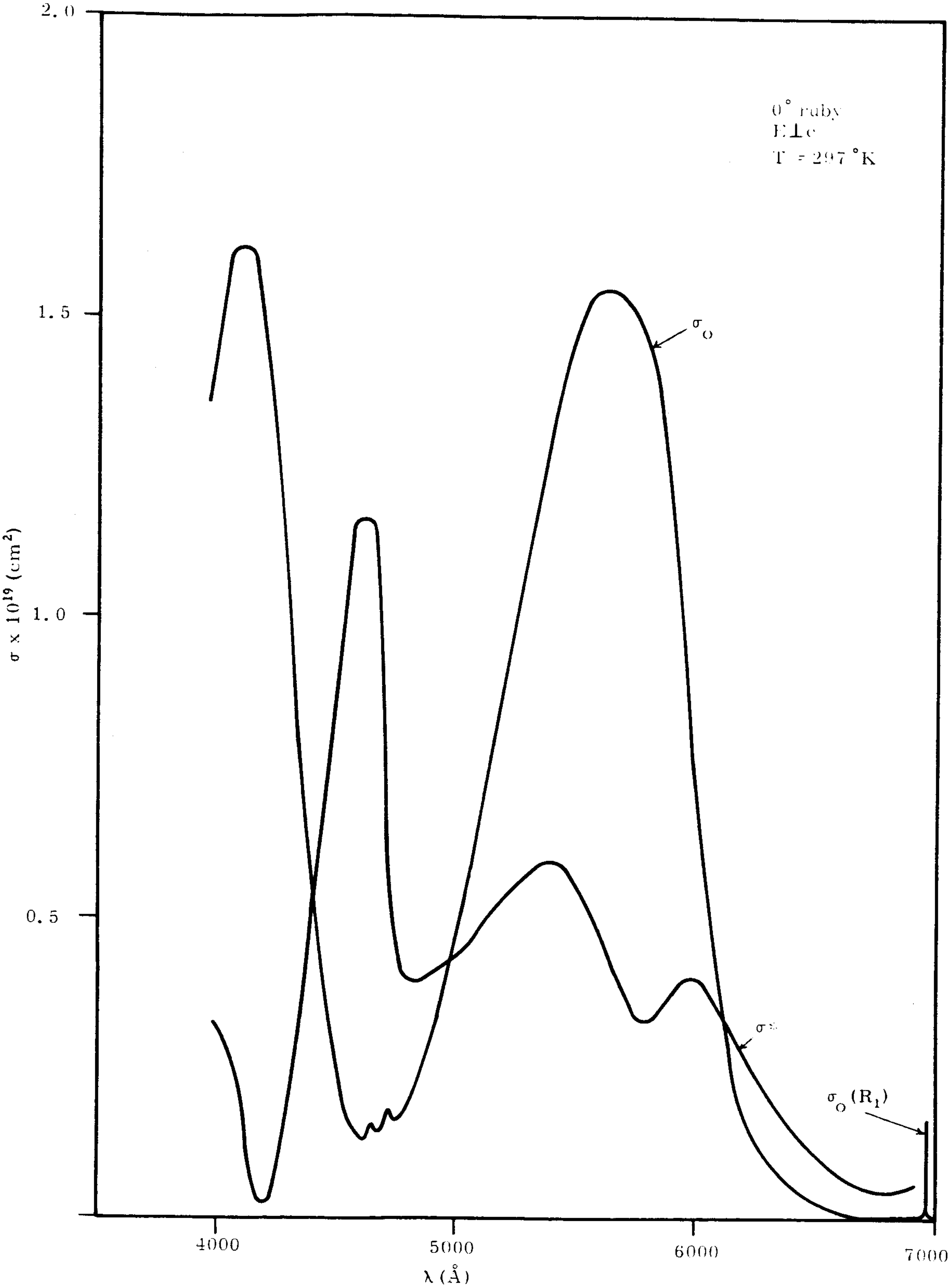

Fig. 1. The ground state absorption spectrum σ_o and excited state absorption spectrum σ^* of Cr^{3+} in ruby at 297 °K.

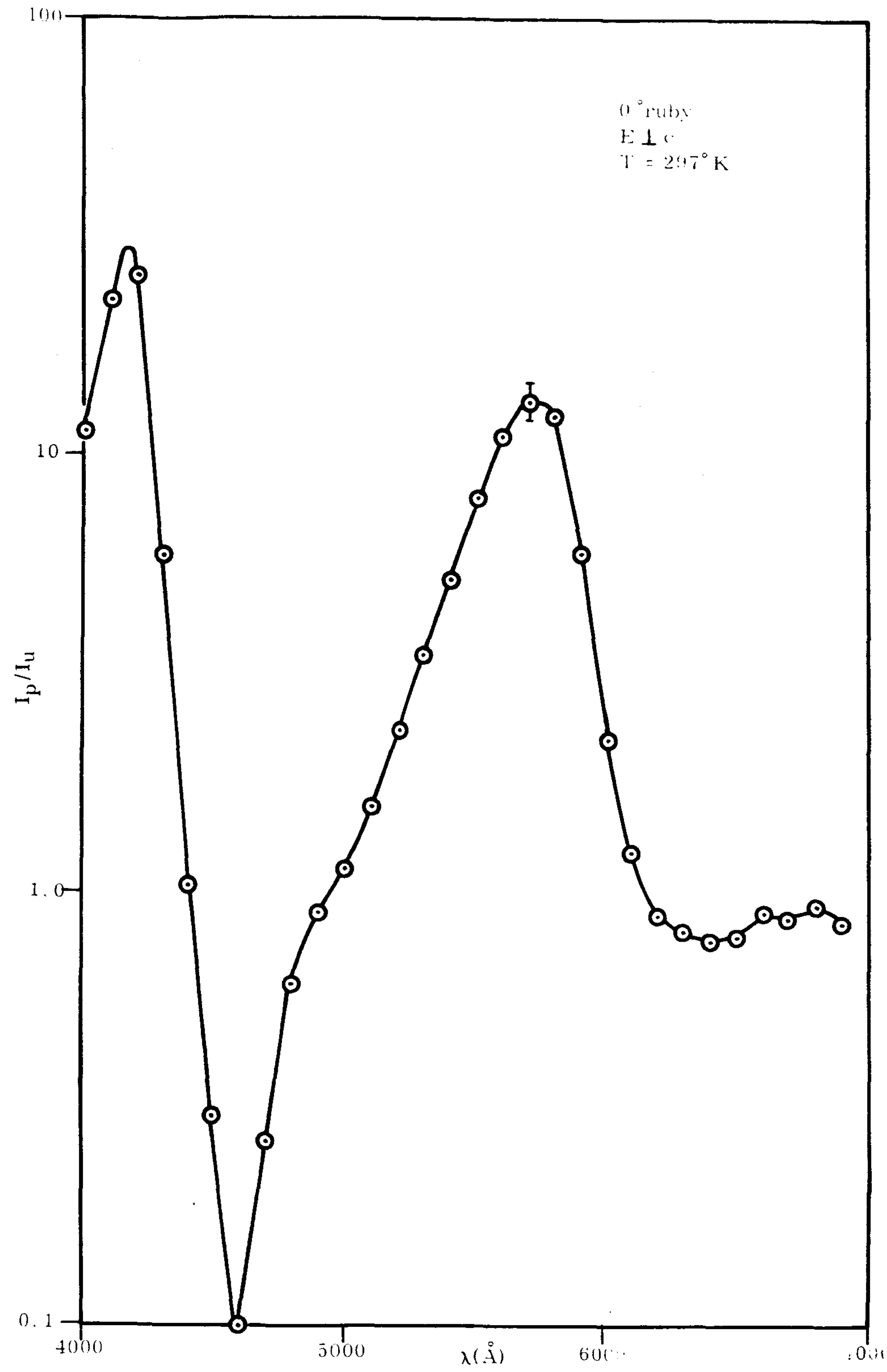

Fig. 2. The ratio of transmitted intensity through the pumped ruby rod I_p to that through the unpumped rod I_u with E ⊥ c at 297 °K.

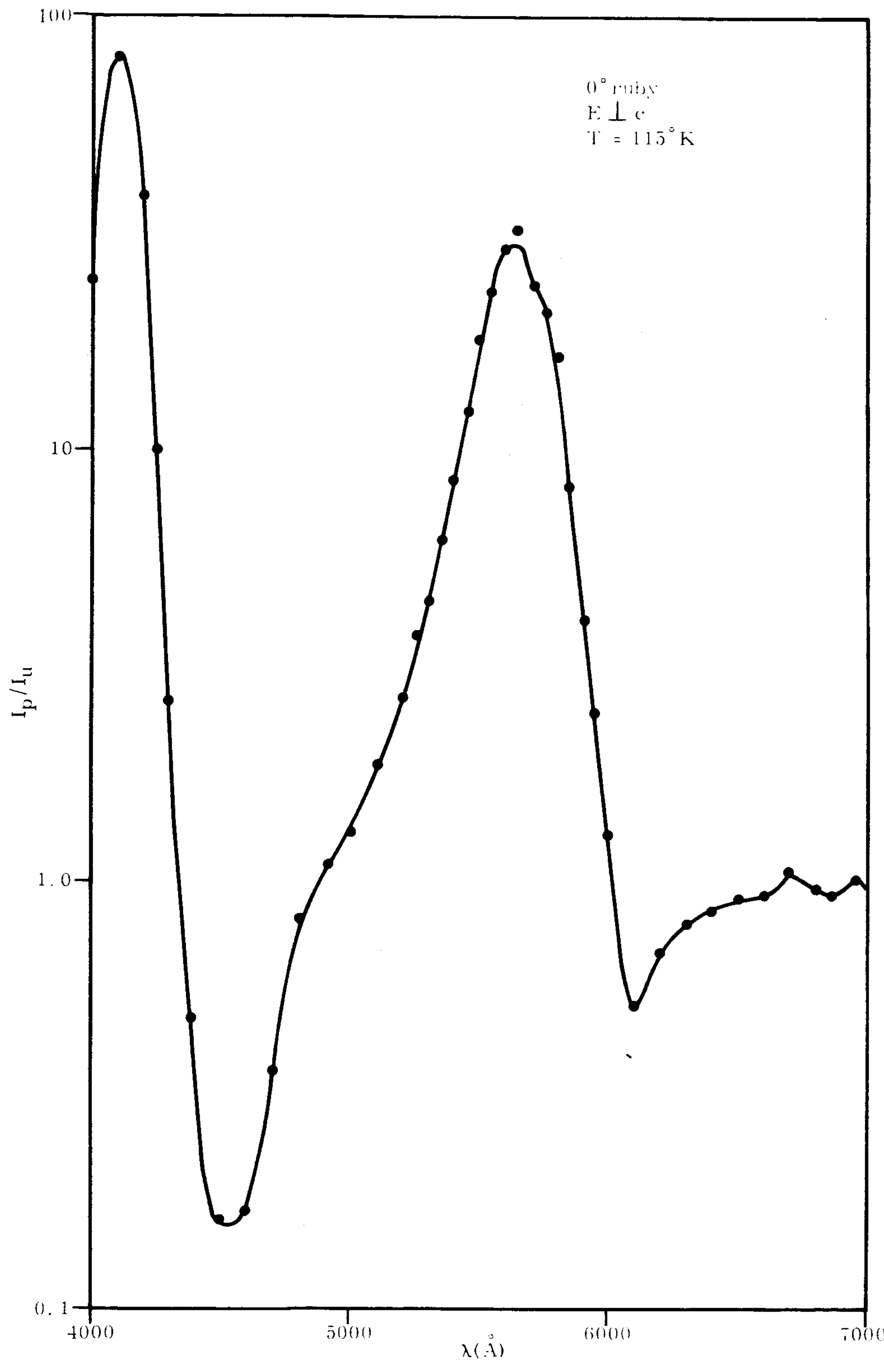

Fig. 3. The ratio I_p/I_u with $E \perp c$ at 115°K.

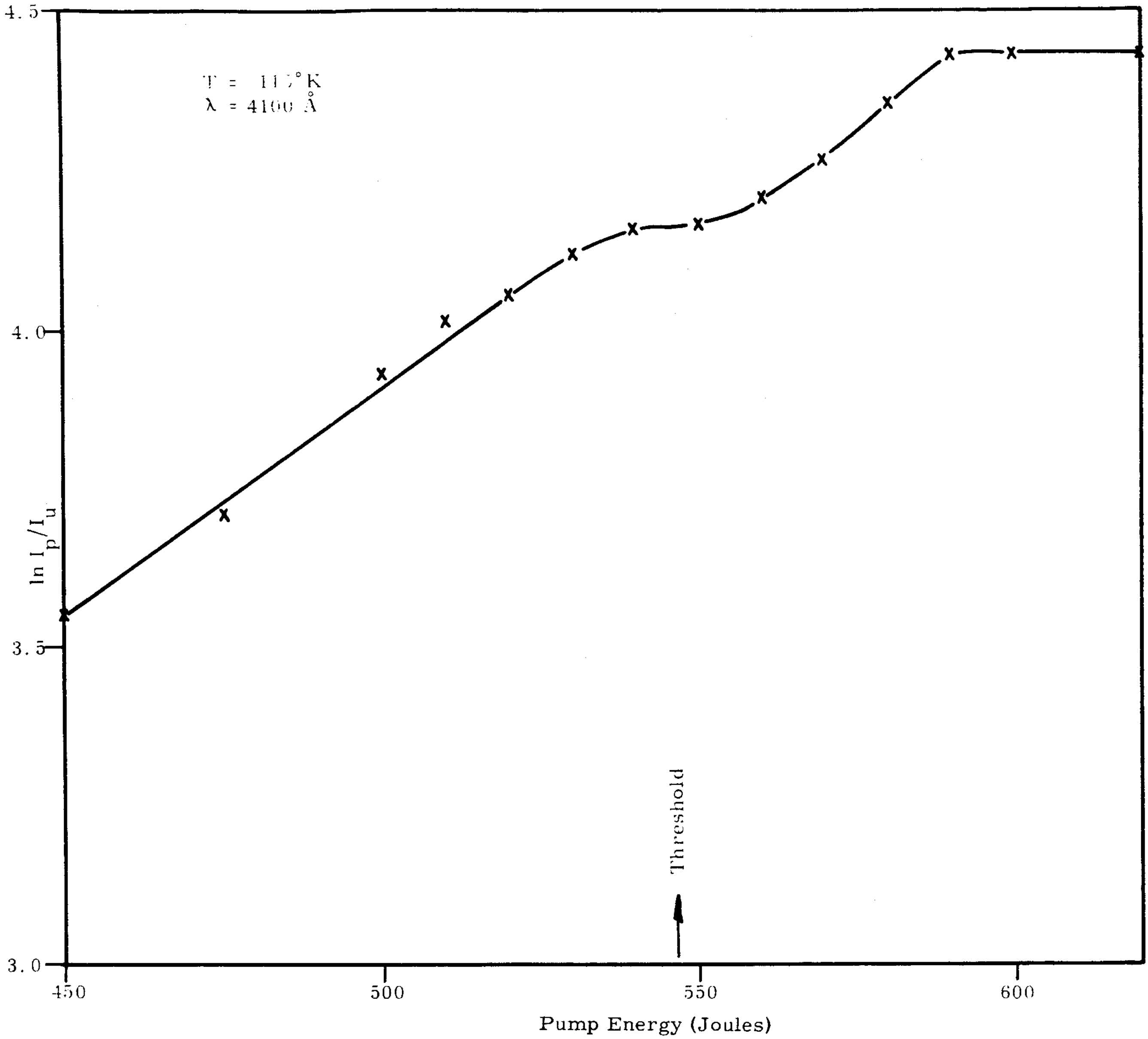

Fig. 4. ln I_p/I_u at λ = 4100 Å as a function of pump energy near laser threshold, probe beam off axis, T = 115°K.

II. STUDY OF YAG: Nd^{3+}

A neodymium doped yttrium aluminum garnet rod 30 mm long by 3mm OD was received from Linde Crystal Products Department at the end of the month. Laser threshold in a FT524 was found to be about 10 joules with a 2 μf capacitor. A typical laser burst observed with an RCA 7102 S-1 photomultiplier is shown in Fig. 5. Fluorescence spectra are being recorded as a function of temperature between 300°K and 100°K. A test of laser quenching in the inhomogeneous magnetic field will be carried out during the coming month.

The observation of time resolved spectra and time resolved radiation patterns has been delayed by the inability of STL Products to provide us with an S-1 image converter camera as scheduled. It appears that the camera will be placed in operating condition early in October, permitting us to proceed with the time resolved studies called for in the contract.

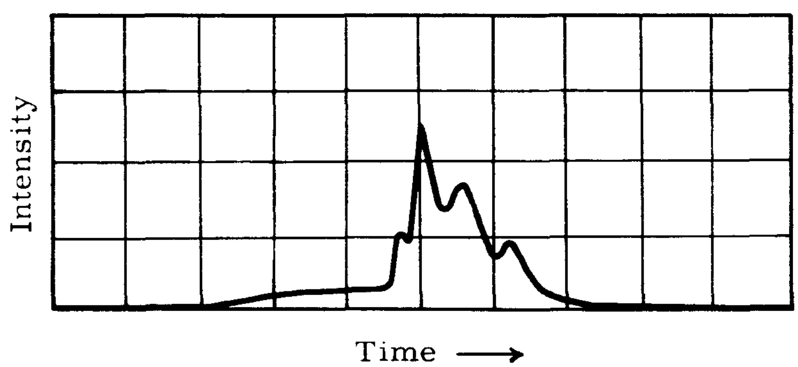

Fig. 5. Laser emission from YAG:Nd^{3+} at 300 °K, 12 joules input to FT 524 flashlamp, sweep speed 10 μsec/div.